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ON THE POSSIBILITY OF HIGH VELOCITY DEFLAGRATION FRONT PROPAGATION BY RADIATIVE ENERGY TRANSFER

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Flash x-ray data of the burning of Hivelite, a proprietary material made by McCormick Selph-Teledyne Company, indicates a "burning" velocity of (about) 383 m/s. Since this exceeds the velocity of deflagration arising from thermal conduction initiation and falls below the value of detonation velocities which arise from shock propagation and thus exceed a material's sound velocity, the existence of a distinct ignition wave front propagation mechanism is to be considered for this material. A new mechanism of ignition front propagation that depends on IR radiation transmission into a highly IR transparent flame sensitive material is proposed and formulated. Present data for the properties of Hivelite are not complete enough to determine if its burning rate arises from an "ignition wave" of the type treated here, but the present data is compatible with that interpretation.

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I. INTRODUCTION

Chemical reaction in propellants and explosives is achieved by heating the constituent molecules to the material's ignition temperature. Heating is achieved directly by contact with other materials at high temperature or indirectly through friction, impact, etc. Once initiated, such chemical reactions can continue to propagate into the bulk material as deflagration or detonation with a more or less well defined wave front. Obviously, the propagation of a deflagration or detonation wave is maintained by the transfer of some of the energy of combustion into the surrounding unburned material which serves to heat the neighboring material to the ignition temperature. Conventional wisdom in physics points out that there are three ways by which thermal energy can be transported - conduction, convection, and radiation. For the present purpose we will distinguish the transfer of kinetic energy in the form of a pressure pulse by a shock wave (or sound wave, as appropriate) from other types of convection mechanisms. The resulting list of mechanisms for energy transfer through a medium is:

1. Conduction.

2. Convection.

3. Shock Wave.

4. Radiation.

It is expected that each of these mechanisms should be encountered, singly or in combination, as the underlying physical process governing propellant and explosive materials.

Conduction is ordinarily a slow mechanism for the transfer of thermal energy. As a consequence, the velocity of a conduction mediated reaction front through a material should be expected to be correspondingly slow. In fact, propellants ordinarily propagate a deflagration front at a velocity of less than about 1 cm/s. Higher values have been reported for certain rocket propellants, but as we will see such materials may belong in a different category.

Convective propagation, the transfer of energy by the bulk motion of the material, can arise in porous media. In the combustion of a confined propellant powder, such as black powder, hot gases at the burning front are forced through the porous medium to ignite the neighboring material. The velocity of the burning front in such a material will correspond to the velocity of the expanding gas that moves through the pores of the material. Such velocities depend on the free expansion velocity of the gases which in turn depends on the sound velocity and ratio of specific heats in the unexpanded gas. Typical velocities are one to several hundred meters per second. Reducing the

size of the pores in the material obviously will reduce the velocity of propagation in such materials. Such a mechanism, therefore, is not to be expected in materials pressed to their maximum or theoretical density tested under high confinement conditions, or conditions precluding explosive confinement interactions as discussed below.

The propagation of a chemical reaction in a material by shock heating is encountered in explosives. The chemical reaction gives rise to a high pressure that drives the shock wave in the explosive material. Heat generated as the shock passing through the material raises the temperature of the explosive to its ignition point to maintain the detonation wave. Since shock velocities exceed the sound velocity in any medium, detonation wave velocities exceed the mediums sound velocity resulting in detonation velocities ranging from 4.9 km/s for baratol to 9.1 km/s for Astrolite. Lower metastable detonation wave velocities, also called low velocity detonation or nondetonative explosions are observed in many liquid and solid explosives[1]. These lower velocities have been attributed[1] to plastic wave propagation in the explosive confinement system. As such, the mechanisms involved do not represent characteristics of the materials alone and lie outside the present discourse.

Thus, we find the first three in the list of mechanisms for energy transfer, conduction, convection, and shock, well represented as important mechanisms determining the ignition propagation velocity regime in bulk materials. The physics of these three has been well discussed in the literature. However, the mechanism of radiative transfer, as should be expected to occur in highly flame sensitive mediums that are transparent to visible or IR radiation, has not been identified or treated previously. Some existing materials such as Hivelite have a burning velocity even in the fully pressed form that appears to lie outside the range of convective deflagration, and yet below the region of shock propagated explosion. The 5A Hivelite composition, tested by A. A. Juhasz of the Ballistic Research Laboratory maintains a high apparent propagation velocity of 300 to 500 m/s up to 98 to 99% of its theoretical density. Such materials may maintain the burning wave front by means of a radiative mechanism.

II. FLASH X-RAY TEST OF BURNING RATE OF HIVELITE COMPOSITION

Flash x-rays have been made to characterize the burning of the Hivelite compound. These flash x-rays were made by Stanley Golaski of the Ballistic Research Laboratory. A 1.27 cm (1/2 inch) diameter 8.89 cm (3.50 inch) long specimen of the Hivelite material mixed with an epoxy binder and pressed into a nonporous solid was inserted into an aluminum cylinder about 10 cm in length and initiated using an electric match. The leads to the electric match can be seen on the left in each of the flash x-rays shown in Figure 1. A 2.38 mm (3/32 inch) thick 1.27 cm (1/2 inch) diameter disc has been attached to the front of the propellant charge. The propellant charge fits loosely into the aluminum tube. The initial configuration can be seen in Figure 1A.

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[1] *Frey, R. B. and Trimble, J. J., "Nondetonative Explosions in Confined Explosive Charges," Seventh Symposium on Detonation, 16-19 June 1981, US Naval Academy, Annapolis, MD.*

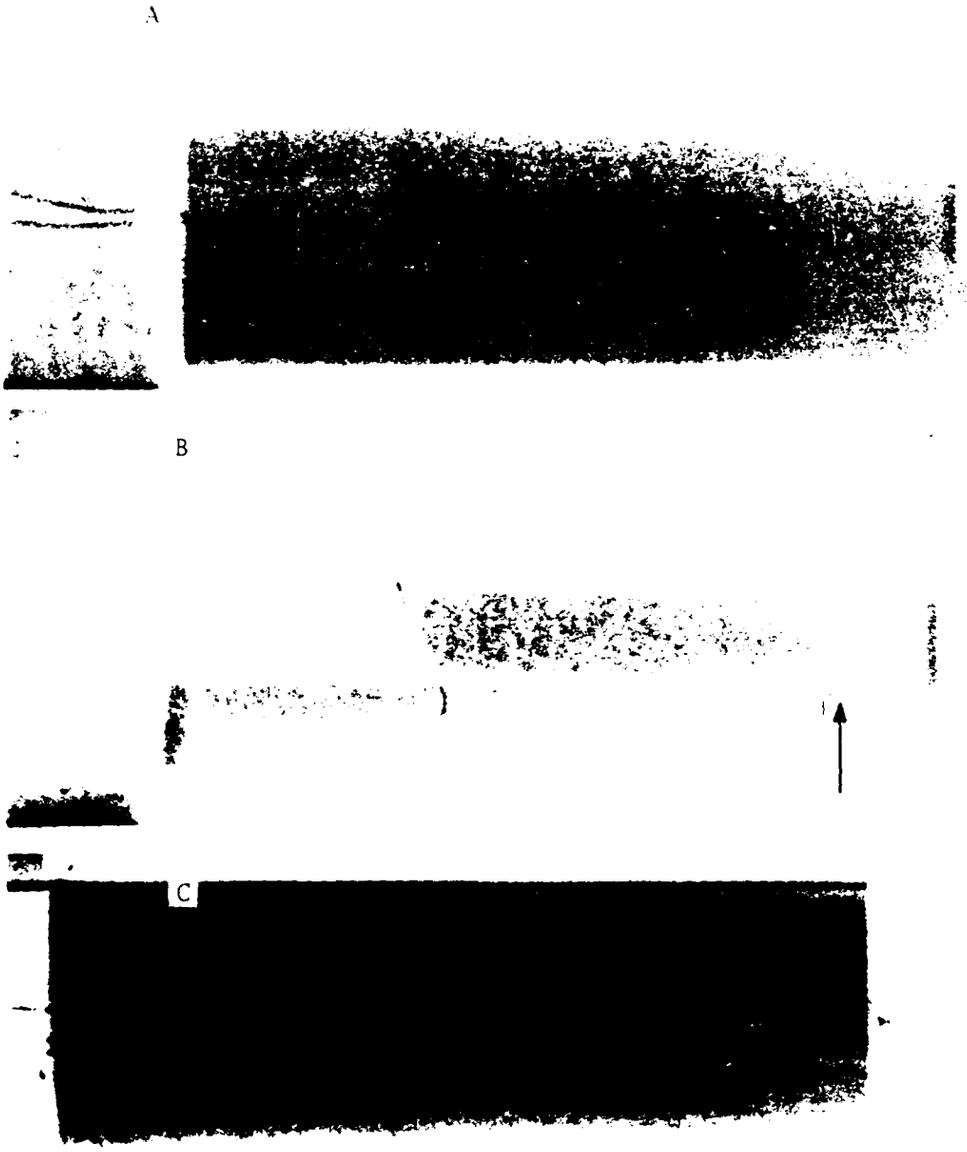

Figure 1 - Flash x-rays of the ignition by electric match of a 1/2" by 3.5" nonporous pressed stick of a Hivelite-epoxy mixture. A 3/32" x 1/2" diameter steel disc is attached to the propellant at right.
A - Shows the initial configuration of the aluminum tube containing the Hivelite rod. The electric match leads can be seen to the left.
B - 84.9 μs after initiation with burning front having traveled 3.25 cm. The pressure front at 7.2 cm can be recognized by the bend (arrow) produced in the originally straight sample.
C - Flash x-ray at 146.7 μs. The three x-rays are views of the same event.

Figure 1B shows the flash x-ray at 84.9 μs after initiation of the electric match. At this time, the ignition front has move 3.25 cm into the Hivelite as indicated by the banding of unburned epoxy and perhaps other constituents of the Hivelite composition. Ignoring the time delay of the electric match, this gives an ignition front velocity W = 383 m/s. This value probably represents a minimum value. Inclusion of 10 μs delay in the electric match would raise the velocity to 434 m/s while the possibility of unresolved burning beyond 3.25 cm would raise the value further. Over estimate should not exceed 5%.

It is also possible to discern in Figure 1B a distinct displacement or bending of the Hivelite rod resulting from the propagation of a pressure pulse into the material. The front of this acoustic wave lies at about 7.2 cm from the initiated end of the rod. This gives an approximate value for the pressure wave velocity of 848 m/s. Since the shape of the wave is continuous and since 848 m/s is too small to indicate any significant particle velocity, we should take this to be the longitudinal sound velocity in the rod rather than a shock wave.

The bending of the rod appears to be related to the fact that the rod is initially tilted at a small angle to the bore of the aluminum tube. The bore is 1.43 cm (9/16 inch) in diameter. This angle results in a component of the force on the rod being directed at an angle of slightly under 1° to the axis of the rod. Thus, the displacement force on a section of length $\Delta\ell$ and radius or from a driving pressure p is

$$F = \pi r^2 \, \Delta\ell \, p \tan \alpha \qquad (1)$$

which can be set equal to the mass $\pi r^2 \Delta\ell \rho$ times the acceleration a, where ρ is the density. This in turn gives the transverse velocity at a time t, simply :

$$V_{\perp} = at \qquad (2)$$

The ratio $V_{\perp}$ to the sound velocity V_s gives the angle Θ of the section of material along the end. Thus,

$$\tan \Theta = V_{\perp} / V_s = \frac{p \, t \tan \alpha}{\rho \, V_s} \qquad (3)$$

from which we can solve for the pressure.

From Figure 1B we obtain at 1 cm (i.e., 12 μs from the wave front) a displacement angle $\Theta \approx 0.7^{\circ}$. This indicates a pressure of about 6.35×10^9 dynes/cm^2 (9.21×10^4 psi). The figure can only be taken as suggestive, however, since we cannot be certain as to the alignment of the shock front driving the rod. Thus, tan α may be significantly larger. For $\alpha = 10^{\circ}$ we would calculate a pressure one tenth as large.

In Figure 1C at t = 146.7 μs, it is seen that the ignition front has propagated entirely through the rod. Material has been driven out the end of the tube. The steel disc has been displaced a distance of 2.8 cm. If we assume an ignition velocity of W= 383 m/s, the disc has an average velocity of 328 m/s or an average acceleration of 106×10^9 cm/5^2. The pressure required to accelerate the disc to this velocity is 1.97×10^9 dynes/cm^2 or 28,600 psi.

Certainly the above pressure estimates are only that, estimates. But they are useful as an aid in determining the nature of mechanisms involved in the burning of this material.

Figures 1B and C also show clearly that a significant portion of the Hivelite composition is not burning, but form solid platelets. The nature of these platelets, which may be epoxy binder, remains to be determined.

III. THEORY

Consider a transparent (or translucent) medium with absorption coefficient A in which a plane wave steady state ignition front is propagating with a velocity W along the x-axis as in Figure 2. If we set the temperature at the ignition front to be T_f, then the energy radiated by the products of the chemical reaction per unit area and time, R_o, will be given by the equation

$$R_o = a\sigma T_f^4 \tag{4}$$

where σ is the Stefan-Boltzmann constant and a is the radiation (absorption) coefficient for the gases. Thus, the flux of radiant energy R at any distance X in front of the ignition wave can be obtained from Equation (4) and the equation for uniform absorption:

$$dR/dx = -AR \tag{5}$$

or integrating

$$R = R_o e^{-Ax} = a\sigma T_f^4 e^{-Ax} \tag{6}$$

Now, if we consider the approach of material toward the ignition front from a frame of reference moving with that ignition front at a velocity W, it will be seen that absorption of energy occurs on the exponential curve, Equation (6), until the material has been raised from its initial temperature T_o to T_{ig}. Thus, we can write, where C_v is the specific heat at constant volume, M the molar weight and ρ the density

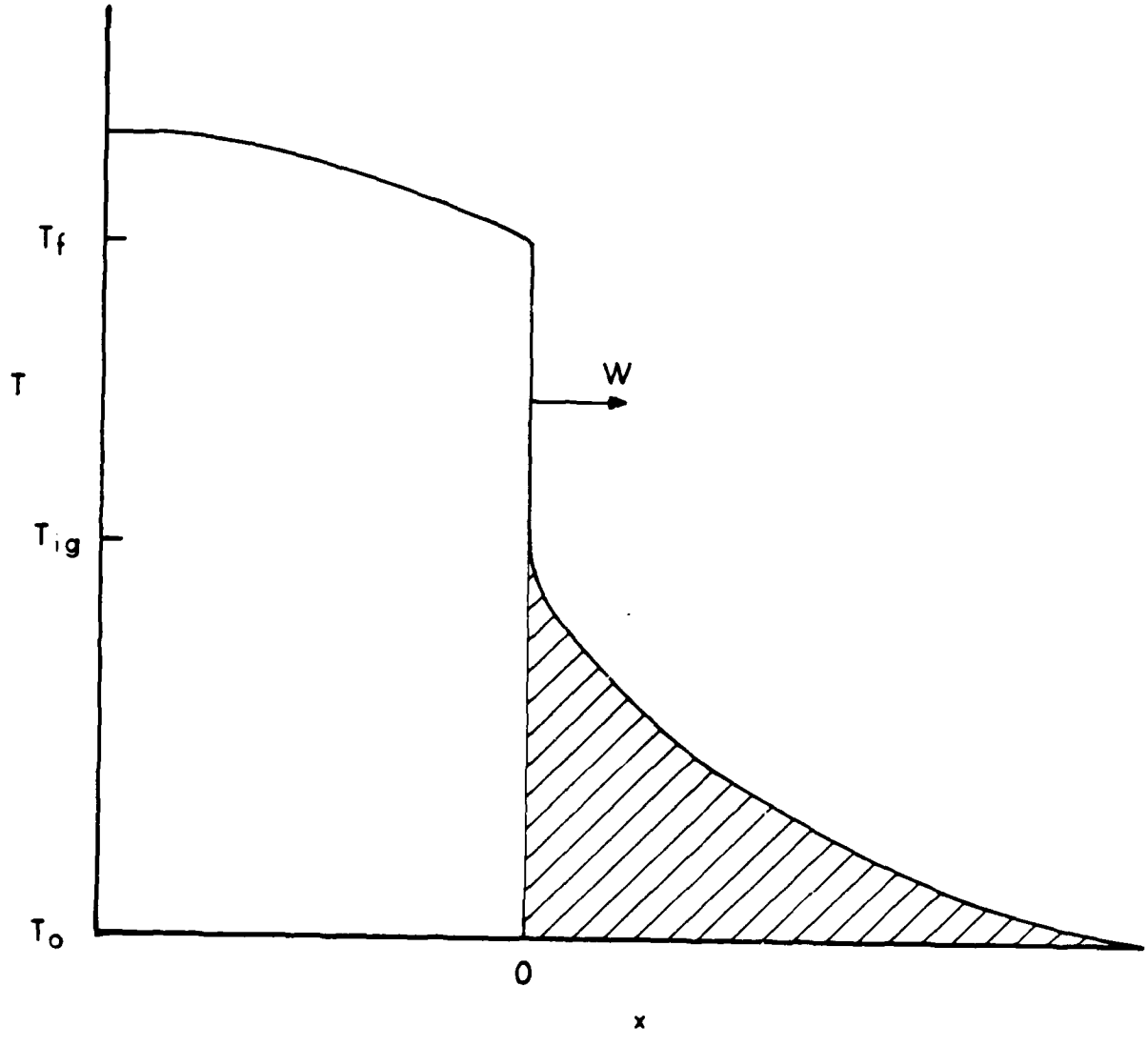

Figure 2 - Schematic for a radiative wave ignition front temperature, T, vs distance, x, graph; T_o, ambient, T_{ig}, ignition, and T_f flame temperatures. Ignition front shown at x=o. The wave front velocity is W.

$$T_{ig} = T_o + \frac{M}{C_v \rho} \int_o^{\infty} (R/W)\, dx \qquad (7)$$

Substituting from Equation (6) and integrating, we have

$$T_{ig} = T_o + \frac{R_o M}{W C_v \rho} \int_o^{\infty} e^{-Ax}\, dx$$

$$= T_o + R_o M / AW C_v \rho \qquad (8)$$

Therefore, the velocity of the ignition front for a radiation ignition wave is

$$W = R_o M / C_v \rho A\ (T_{ig} - T_o) \qquad (9)$$

or interms of the temperature of the ignition front

$$W = a M \sigma\ T_f^4 / C_v \rho A\ (T_{ig} - T_o) \qquad (10)$$

From Equation (10), we see that the radiative ignition wave velocity depends principally on the existence of a very small absorption coefficient coupled with a great flame sensitivity (small value of the ignition temperature above ambient). A small specific heat at constant volume and a high combustion temperature are additional important factors for radiative ignition wave propagation. It should be noticed that a simple characteristic of the radiative ignition wave that can be used to discriminate this form of ignition wave from other ignition mechanisms is the direct dependence of the wave velocity on the materials ambient temperature.

IV. IR ABSORPTION IN HIVELITE

An IR spectrum for the Hivelite compound was obtained by Arpad Juhasz, Applied Ballistics Branch, Interior Ballistics Division, Ballistic Research Laboratory, using a Perkin Elmer 237B Infrared Spectrophotometer. The sample was mixed with KBr as matrix and pressed into a 3/4 cm pellet. The spectrum shows a principle absorption line at 1380 cm^{-1}, a sharp medium line at 850 cm^{-1}, a sharp weak line at 1770 cm^{-1}, a medium absorption line at 2450 cm^{-1}, a weak absorption line at 2900 cm^{-1}, and a broad weak absorption at 3400 cm^{-1}. The transmission in the other

regions of the IR was high. Generally, the absorption in the IR is not extensive or great. The material is translucent in the IR, but an estimate of the absorption per cm has not been obtained. One can only state at present that the available data is consistent with high IR transmission and the lack of extensive IR absorption structure supports this view.

The flame temperature of the Hivelite has been estimated by Arpad Juhasz to be about 2800°K and the ignition temperature to be in the 400°K to 500°K range*.

V. ESTIMATES OF IGNITION WAVE VELOCITY

The present data on Hivelite is not adequate to allow evaluation of Equation (10). Approximate numbers are available that will give an indication as to the possible utility of Equation (10). The radiative coefficient a can be taken to be unity, the molecular weight is known to be about 100, the temperature at the ignition front is about $T_f = 2800^{\circ}K$, density of the fully pressed pure material about $2 g/cm^3$, and the specific heat is in the range of 0.3 cal/mole°K. For $T_{ig} = 400^{\circ}K$, we obtain $W = 1.39/A$ (cm/s). For absorption coefficients of $A = 0.05\ cm^{-1}$, $0.02\ cm^{-1}$, and $0.01\ cm^{-1}$ we obtain W = 27.7 m/s, 69.3 m/s, 138.6 m/s. These values are considerably larger than those for conduction deflagration. The values are not as high as our estimate of 383 m/s for the ignition wave velocity in Hivelite obtained from the flash x-rays. Nevertheless, the values are sufficiently close to lend support to the position that the mechanism operating in the ignition of Hivelite is that of an IR ignition wave.

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*It may be noted in this connection that the principle commercial use of Hivelite has been in the manufacture of energy transfer lines to active explosive bolt release mechanisms in missiles and in military aircraft ejection systems (as in the F111 aircraft). A principle advantage Hivelite is reputed to have according to the manufacturer over other systems such as electrical explosive bolt activating lines is that the great flame sensitivity allows the transfer line to function even when extensively damaged, as for example, if several inches of the line have been physically removed or breached. High sensitivity to flame initiation is a characteristic of this material.

VI. SUMMARY AND CONCLUSIONS

Flash x-rays of the ignition and burn of Hivelite show that material has a burning rate of about 383 m/s and propagates a high pressure wave at a velocity of about 848 m/s (error bars on these values may range from -5% to + 20%). The x-rays indicate that the material's burning mechanism does not arise from conduction or shock mechanisms. A new "ignition wave" mechanism has been proposed and formulated that within the present limits of available information is compatible with the observed high burning rate of Hivelite.

The present data clearly indicates the need for extensive flash x-ray studies on the Hivelite and similar propellant materials to obtain further data on propagation velocity, pressure wave velocity, pressures, and propellant-projectile interaction.

In addition, data should be obtained to determine flame sensitivity, IR transmission, specific heat, and flame temperature. Since the theoretical equation for the propagation velocity indicates a strong dependence of the ignition wave velocity on the ambient temperature, tests should be conducted to look for this dependence.

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